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Process for producing an aryl imido-peralkanoic acid.

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Process for producing an aryl-imido-peralkanoic acid by means of the peroxidation with H_2O_2 of the corresponding alkanolic (non-peroxy) acid, in the presence of a strong acid with a pKa value equal to, or lower than 3, and in the presence of a halogenated solvent, characterized in that said solvent is CH_2Cl_2 or $CHCl_3$, that the amount of alkanolic acid submitted to peroxidation is equal to or smaller than the solubility threshold of said alkanolic acid (in said solvent) and that a solution of said acid in said solvent is reacted with H_2O_2 (at 10-35°C), with the molar ratio of said strong acid to said alkanolic acid being lower than 2.

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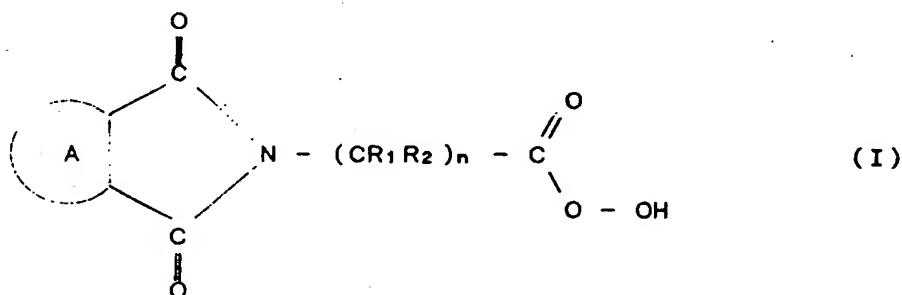
The present invention relates to a process for producing, in continuous mode, an arylimidoperalkanoic acid, in particular ϵ -phthalimidoperhexanoic acid. ϵ -phthalimidoperhexanoic acid -- designated in the following as "phthalimidoperhexanoic acid" or, more briefly, "PAP" -- is used as a bleaching agent in low-temperature washing of textile products, as reported in European patent 325288, and is respectively used as well as a bleaching agent, a sanitizer, an oxidizer agent and a free-radical generator, in the processing and treatment of textile products, paper and wastes, and in hygienic treatments and in chemistry (of intermediates and polymers).

From U.S. patent 4 172 086, the contents of which are an integral part of the instant specification, producing peralkanoic acids (in particular peralkanedioic acids) by oxidizing the corresponding alkanolic (non-peroxy) acids with hydrogen peroxide and in the presence of large amounts of H_2SO_4 or oleum, by feeding to the reaction zone the acid to be peroxidized, as a suspension (in halogenated organic solvents), is known; unfortunately, the amounts of H_2SO_4 were rather large (preferably of from 2 to 3 mol per mol of alkanolic acid) and the necessary temperatures for peroxidizing the high-molecular-acids ($C_{10}+$) were also rather high ($40-60^\circ C$).

Furthermore, when the present Applicant tried to cause the suspension of a phthalimidoalkanoic acid in some of the halogenated solvents mentioned by said patent (carbon tetrachloride, monochlorobenzene, and so forth) to react by a continuous process, the test had to be discontinued after a few minutes owing to the clogging of the equipment, caused by an enormous increase in reaction mixture viscosity (see Comparative Example No. 4).

The same Applicant developed now a novel process which makes it possible arylimidoalkanoic acids to be peroxidized at low temperature and on a large scale, in continuous mode and with flexibility, in compliance with the safety requisites and with very reduced aliquots of reactants, additives and auxiliary services.

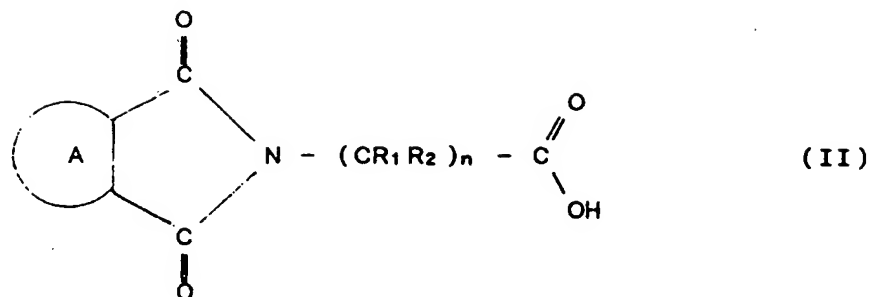
In its widest aspect, the present invention relates to a process for producing in continuous mode an arylimidoperalkanoic acid having formula (I):



in which n is an integer comprised within the range of from 3 to 7 and:

- A is a benzene or naphthalene ring, optionally bearing one or more substituent groups, in particular bearing a carboxy group;
- R_1 and R_2 , which can be either equal to, or different from, each other, are selected from among H and the linear or branched, possibly substituted, alkyl groups of from 1 to 5 carbon atoms;

by means of the peroxidation with H_2O_2 of the corresponding arylimidoalkanoic (non-peroxy) acid of formula II:



in the presence of a strong acid with a pKa value equal to, or lower than, 3 and preferably equal to, or lower than, 1, and in the presence of a halogenated organic solvent, characterized in that:

(A) said halogenated solvent is selected from among methylene chloride (CH_2Cl_2) and trichloromethane (CHCl_3);

(B) a solution is prepared of the arylimidoalkanoic acid to be peroxidized (i.e., the raw material) in said halogenated solvent, by taking care that the amount of raw material dissolved is:

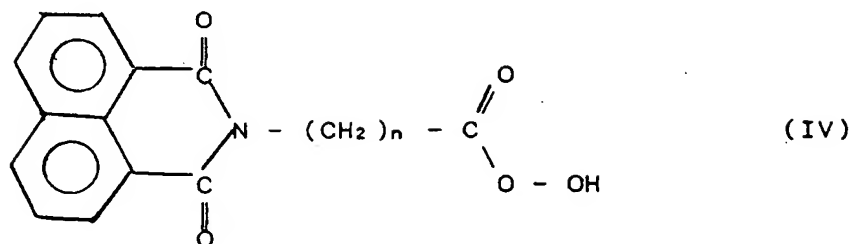
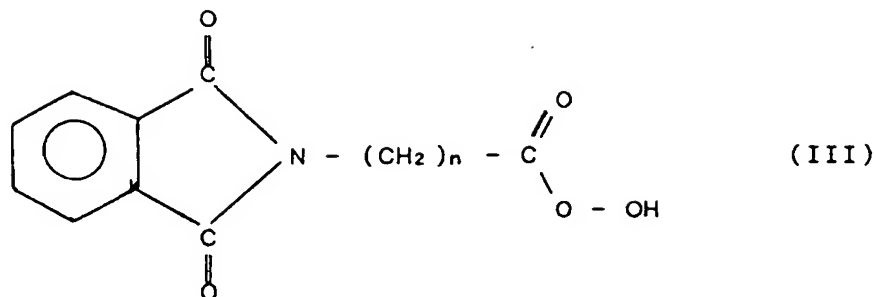
(i) either equal to, or smaller than, the solubility threshold of said raw material in said solvent at the reaction temperature as defined hereinunder in (C);

(ii) higher than that amount of raw material which stoichiometrically corresponds to the solubility threshold of said peralkanoic acid in said solvent [at the same temperature as defined in above (ii) item];

(C) the solution obtained according to (B) is continuously reacted, at $10-35^\circ\text{C}$, with aqueous H_2O_2 , in the presence of said strong acid, with the molar feed ratio of said strong acid to the acid to be peroxidized being adjusted at a value smaller than 2 (preferably smaller than 1.3), with a reaction product constituted by two liquid phases (an aqueous phase and an organic phase) being thereby obtained;

(D) said reaction product is separated into two component single homogeneous phases, said peralkanoic acid and halogenated solvent are recovered from the organic phase and said halogenated solvent is recycled to the dissolving step as described in (B).

In particular, R_1 and R_2 are selected from among H and C_1-C_5 alkyl groups bearing carboxy, percarboxy, hydroxy (OH), nitro (NO_2) or C_1-C_5 alkoxy substituents. Examples of peracids which can be obtained according to the invention are phthalimidoperalkanoic acids of formula (III) and 1,8-naphthaleneimidoperalkanoic acids of formula (IV) (a particularly interesting acid is ϵ -phthalimidoperhexanoic acid):



25 The selection of the solvent and of the concentration values is critical; the present Applicant tried to use very similar solvents from the chemical point of view, such as carbon tetrachloride, monochlorobenzene or acetonitrile, always meeting with serious drawbacks, which are discussed in greater detail in the following.

30 Going into detail, the present Applicant could unexpectedly observe that not only that particular peroxy product (PAP), but also the particular raw material (PAC) is -- contrarily to the teaching of the prior art -- soluble, and even more soluble, in said particular halogenated solvents; thus allowing the reaction to proceed according to a different mechanism type (relatively to as disclosed in U.S. patent 4 172 086) and the novel mechanism to lead to high conversion and yield values under low temperature, low viscosity and internal system temperature control conditions. In the mean time, a very efficient product recovery can be accomplished (also from the inorganic phase). Of course, the pressure should be such as to keep the system in the liquid phase. Table 1 shows some solubility data experimentally observed by the present

35 Applicant.

Table 1 (*)

Solvent	Solubility (g per 100 cm ³ of solvent)	
	PAC	PAP
Dichloromethane		
at 14°C:	51.1	26.3
at 25°C:	67.3	35.2(**)
Trichloromethane		
at 14°C:	52.3	26.1
at 25°C:	67.9	34.9

(*) See also chart of Figure 3.

(**) Estimated value

The new process makes it possible the reaction of peroxidation to be carried out in continuous under controlled and safe conditions, with very low temperatures and very low aliquots of strong acid and hydrogen peroxide, by preferably using mixing reactors of CSTR (Continuous Stirred Tank Reactor) type, with flexible size and potentiality and useable at the commercial level. As an alternative, an external-recycle reactor or a static mixer might be used; furthermore, the feed of the reactants may be gradual, i.e., can it can be accomplished at a plurality of successive feed points. These reactors and these operating methods are disclosed, e.g., in European patent 0970, the contents of which constitute an integral part of the present specification. Cheapness, flexibility and possibility of reaching considerable scale increases (in compliance with the safety requisites) are qualifying features of the process according to the invention.

According to a particularly advantageous form of practical embodiment of the invention, the adoption of the following, either complementary or alternative, operating instructions, is recommended for peroxidizing PAC:

- continuous feed of a solution containing at least 20% (preferably 25-30%) by weight of acid to be peroxidized (in particular, PAC in a halogenated solvent);
- continuous feed of aqueous hydrogen peroxide at 50-90% by weight (preferably 60-80%) and with a molar ratio of H₂O₂ (100%) to PAC of from 1 to 2 (preferably of from 1.01 to 1.6);
- molar ratio of the strong acid to PAC comprised within the range of from 0.50 to 1.29; while said strong acid can be selected, e.g., from among sulfuric acid, oleum, methanesulfonic acid, trifluoromethanesulfonic acid, fluorosulfonic acid and their mixtures; and so forth.

The amount of strong acid can be considerably reduced if, before (or during) the contact with the strong acid, a so-called "solid acid" is used; with said solid acid being selected from the group consisting of the cation exchange resins, of strong type, and the zeolite aluminosilicates, of PENTASIL type (in their acidic form; e.g., HZSM5 and HZSM11). In other terms, the contact with a bed of said resins or said aluminosilicates can reduce the value of the ratio of the strong acid to PAC to be peroxidized, even down to very low levels (0.50, by mol).

At the end of the peroxidation, in one or more reactors (in parallel, or, preferably, in cascade), the double-phase reaction mixture must be separated (phase separation), e.g., in a separation tank; the acidic/aqueous phase can be subsequently diluted with deionized water, in order to facilitate the recovery of the residual product (PAP) from said acidic/aqueous phase, and reduce the risk of instability thereof. The amount of dilution water is generally comprised within the range of from 0.5 to 1.5 litres per each litre of separated aqueous phase. As an alternative, the dilution can be performed before carrying out the phase separation. The organic phase, containing traces of residual acidity, is usually neutralized with a slightly alkaline solutions, containing, e.g.: NaHCO₃, Mg(HCO₃)₂, NaOH, and so forth; as an alternative, said organic

phase can be caused to flow through a bed of anion exchange resins, or of solid alkali-metal compounds or alkali-earth metal compounds (NaHCO_3 , Na_2CO_3 , MgCO_3 , and so forth).

In the following, a particular form of practical embodiment of the process according to the invention is disclosed with the aid of Figure 1; of course, both the description and the figure are supplied for merely illustrative purposes, and in no way should they be construed as being limitative of the scope of the invention.

Viewing at Figure 1, a concentrated solution of ϵ -phthalimido-hexanoic acid (PAC) in a halogenated solvent (1), aqueous hydrogen peroxide (2) and the strong acid (3), e.g., H_2SO_4 , are fed to reactor "R1", kept at room temperature. In that way, a system is obtained, which consists of two liquid phases. Such phases constitute the reactant system, characterized by low viscosity values and ease of fluid-dynamic dispersion; the normal stirring systems can thus secure exchange surface areas per volume unit, which are large enough as not to confine the specific production capacity of the reactant system within a limiting kinetic control by the phase transfer. Furthermore, such a system is characterized by a high capability of self-control of possible local heat accumulations, deriving from the instability and degradation of reactants and products (the mass is dispersed throughout the reactor as a continuous fluid, without stagnation pockets due to viscosity gradients, and without any local accumulations of solid phase, and is anyway thermally buffered by an excess of low-boiling solvent). Said system is furthermore characterized by an easy processability, in that its phases can be easily separated, by simple decantation (under rest conditions).

The reactant mixture (4) enters a second reactor R2 similar to the first reactor "R1". Inside reactor "R2" peroxidation is completed. The ultimate effluent (5) is submitted to phase-separation inside the separation tank "D". The acidic/aqueous phase (6) is then moderately diluted with deionized water (7), for safety reasons, and in order to enable that aliquot of product (PAP) which is not dissolved in the organic phase to be effectively recovered by back-extraction with the reaction solvent. The diluted solution (8) can be stored in the tank "S", or it can be directly sent to the back-extraction. The organic phase (9), in which traces of residual acidity exist, is neutralized with a weakly alkaline solution (10); as an alternative, the organic phase can be neutralized, as already said, by being caused to flow through a solid neutralizing bed. From the organic PAP solution (11), the product can be easily separated with the required purity level, e.g., by crystallization of the solute (by taking advantage of the considerable temperature dependence of its solubility in the solvent), and therefore it can be recovered by filtration (and/or centrifugation) and drying. According to an alternative route, as said in the following, the product (PAP) can be recovered by distilling off the solvent.

Another form of practical embodiment of the process according to the present invention is depicted in Figure 2

Viewing at said Figure 2, the reaction effluent (5) is phase-separated in an "SM" equipment piece, from which an aqueous phase (12) and an organic phase (13) outflow; a deionized water stream (14) is added, for such purposes as cited hereinabove, to the feed stream (5), and/or to the aqueous phase (12). The organic phase (13) is sent to a recovery section "RC", in which it is preferably admixed with the salt of an alkali-earth metal (15), e.g., magnesium sulfate (MgSO_4), and in which the following are recovered:

(a) the peracid (16), as solid particles, or as a concentrated slurry;

(b) the solvent (17), which is recycled to the reaction zone (peroxidation). The recovery of the solvent can take place in several ways. For example, one might drastically cool the mixture and then carry out a filtration (and/or a centrifugation), or the solvent can be removed by distillation. Said distillation can be performed in vacuum, or it can be a steam distillation, and so forth; see, e.g., the steam stream (18) shown in the figure.

The solvent, whichever is the way it is recovered by, can be fully recycled.

The following examples are supplied for merely illustrative purposes, and in no way should they be constructed as being limitative of the scope of the invention.

In all continuous-run examples, the following equipment was used:

- vertical glass reactors, each one of 1.8 litres of capacity, with variable discharge level, equipped with jacket, flanged head, propeller stirrer (with 6 blades revolving at 650 rpm) and baffles (4 elements);
- a horizontal separation tank (phase separation unit) made from glass, of 0.7 litres of capacity, provided with a storage well and a partition wall.

Example 1

0.42 kg/hour of H_2SO_4 (at 96% by weight), 0.23 kg/hour of H_2O_2 (at 70% by weight) and 4.17 kg/hour of a PAC solution (at 20% by weight) in methylene chloride were simultaneously and in continuous charged to the facility depicted in Figure 1. The reaction temperature was kept at 25°C by cooling with cold water

flowing through the reactor jacket. In that way, after phase separation, two liquid phases, i.e., a heavy, acidic phase (0.68 kg/hour) and a light organic phase (4.14 kg/hour) were obtained inside the "D" tank. The organic phase was continuously neutralized with an aqueous solution of NaHCO_3 and Na_2SO_4 . After phase separation and crystallization, 0.79 kg/hour was obtained of ϵ -phthalimidoperhexanoic acid (PAP) with iodometric titre higher than 99%, and a yield, relatively to the reactant (PAC), of 88%.

Example 2

7.09 kg/hour of a PAC solution (at 23.5%) in CHCl_3 , 0.83 kg/hour of H_2SO_4 (at 96%) and 0.47 kg/hour of H_2O_2 (at 70%) were fed to the same facility as of Example 1. The reaction temperature was kept at 25°C by cooling with cold water flowing through the reactor jacket. After phase separation, the residual mineral acidity was removed by filtering the product solution (in CHCl_3) on a bed of solid MgCO_3 . The product was then separated from the solvent by crystallization, by cooling, and, after filtration and drying, 1.57 kg/hour was obtained of PAP with iodometric titre higher than 99%. The overall yield, relatively to PAC (excluding recycles), was of 89%.

Example 3 (Comparative Example: Batchwise Test with CCl_4)

300 g of CCl_4 , 70.4 g of PAC and 35 g of H_2SO_4 (at 96%) were charged to a calorimetric reactor of 500 cm^3 of capacity, equipped with mobile-blade stirrer and thermometer, dipped in a circulating water bath at 25°C. The mixture was heterogeneous, with undissolved PAC in suspension; said mixture was stirred at 25°C and 19.5 g of H_2O_2 (at 70%) was added to it. After 30 minutes, with stirring being continued, still at 25°C, the reaction mixture, which was heterogeneous and paste-like, with a large amount of suspended product, was discharged into 500 cm^3 of cold, deionized water (5°C) and filtered, with great difficulty, through a porous frit filter and under vacuum. The isolated product was subsequently slurried in 100 cm^3 of deionized water and was neutralized, to pH 6, with strong stirring, with an aqueous solution of Na_2CO_3 (at 10%). The neutralized product was filtered once more and was dried at 25°C for 48 hours (under vacuum) in a CaCl_2 containing drier. In that way, 62 g was obtained of a waxy product containing 62% of PAP (by iodometric assay) and with a yield of 51.7% (relatively to PAP).

Example 4 (Comparative Example: Continuous Test with CCl_4)

The test of Example 3 was repeated, but operating continuously in a facility of the same type as described in Example 1 and Figure 1. The result, which was clearly negative, reported in Table 2, shows once more that only a very limited number of halogenated compounds (CH_2Cl_2 and CHCl_3) can lead to satisfactory results, whilst even slightly different compounds, such as CCl_4 , are decidedly excluded from commercial use.

Example 5 (Comparative Example:

Continuous Test with a PAC Excess "Suspended" in CH_2Cl_2)

The test of Example 1 was repeated by continuously feeding 2.08 kg/hour of a slurry containing a PAC amount much larger than the reactant solubility threshold (approximately 89 g per each 100 cm^3 of methylene chloride). The temperature was kept at 25°C by cooling with water flowing through the reactor jacket; as in Example 4, the test had to be discontinued after about 10 minutes, owing to equipment clogging.

Example 6 (Comparative Example:

Test at a Too High Temperature)

The test of Example 2 was repeated, with the reaction temperature being increased up to 60-65°C, by means of a controlled heating with hot water flowing through the reactor jacket. 1.40 kg/hour of PAP was obtained, with a iodometric titre of 90.4%; the yield, relatively to PAC (excluding the recycles) was of only 71.6%.

Example 7

Example 1 was repeated at a temperature of 30°C. by lowering the H₂SO₄ (100%)/PAC molar ratio to the value of 1,200. A 86% yield was obtained, as reported on Table 2.

Example 8

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Example 7 was repeated by still lowering the H₂SO₄ (100%)/PAC molar ratio to the value of 1,000. A 79% yield was thus obtained, as reported on Table 2.

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Table 2

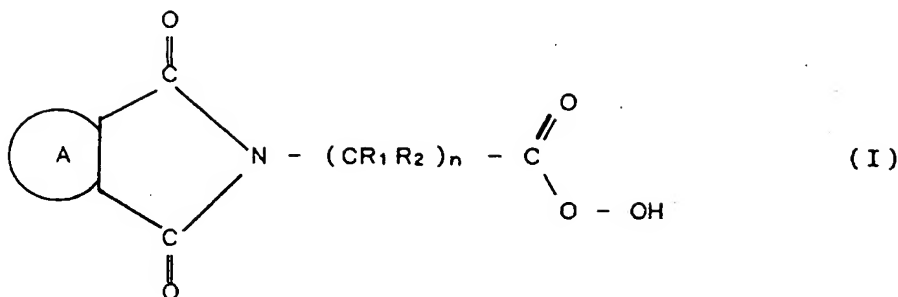
Example	Process Type	Ratio of H ₂ SO ₄ (100%) to PAC (by mol)	Ratio of H ₂ O ₂ (100%) to PAC (by mol)	Halogenated Organic Solvent	Ratio of PAC to Solvent (by weight)	T (°C)	Residence Time	Yield
1	Continuous	1.288	1.48	CH ₂ Cl ₂	20:80	about 25	30 min	88
2	Continuous	1.274	1.52	CHCl ₃	23.5:76.5	about 25	30 min	89
3 ¹	Batchwise	1.270	1.49	CCl ₄	19:81	about 25	30 min	51.7
4 ¹	Continuous	1.270	1.49	CCl ₄	19:81	about 25	-	2
5 ¹	Continuous	1.288	1.48	CH ₂ Cl ₂	89 g of PAC per 100 cc of solvent	about 25	-	2
6 ¹	Continuous	1.274	1.52	CHCl ₃	23.5:76.5	60-65	30 min	71.6
7	Continuous	1.200	1.48	CH ₂ Cl ₂	20:80	30	30 min	86
8	Continuous	1.000	1.48	CH ₂ Cl ₂	20:80	30	30 min	79

¹ Comparative Example.

² Test discontinued after approximately 10 minutes, owing to equipment clogging, in particular due to clogging of connection pipe between reactors R1 and R2.

55 Claims

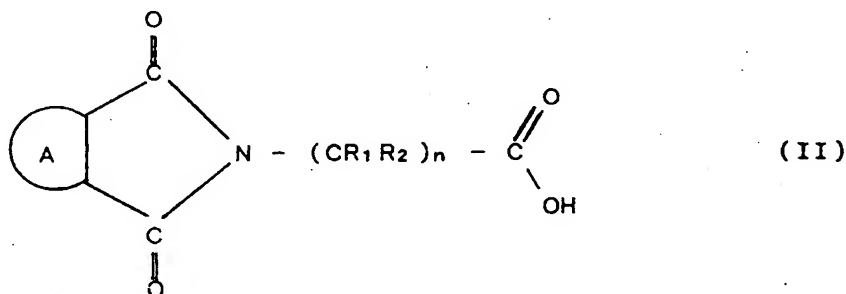
1. Process for producing in continuous mode an arylimidoperalkanoic acid having formula (I):



in which n is an integer comprised within the range of from 3 to 7 and:

- 15
- A is a benzene or naphthalene ring, optionally bearing one or more substituent groups, in particular bearing a carboxy group;
 - R₁ and R₂, which can be either equal to, or different from, each other, are selected from among H and the linear or branched, possibly substituted, alkyl groups of from 1 to 5 carbon atoms;

20 by means of the peroxidation with H₂O₂ of the corresponding arylimidoalkanoic (non-peroxy) acid of formula II:



in the presence of a strong acid with a pK_a value equal to, or lower than, 3 and preferably equal to, or lower than, 1, and in the presence of a halogenated organic solvent, characterized in that:

40 (A) said halogenated solvent is selected from among methylene chloride (CH₂Cl₂) and trichloromethane (CHCl₃);

(B) a solution is prepared of the raw material arylimidoalkanoic acid to be peroxidized in said halogenated solvent, caring that the amount of raw material dissolved is:

(i) either equal to, or smaller than, the solubility threshold of said raw material in said solvent at the reaction temperature as defined hereinunder in (C);

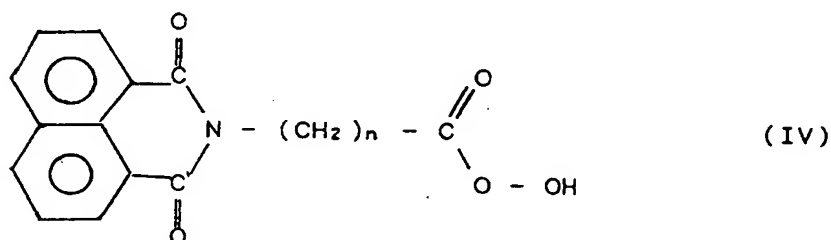
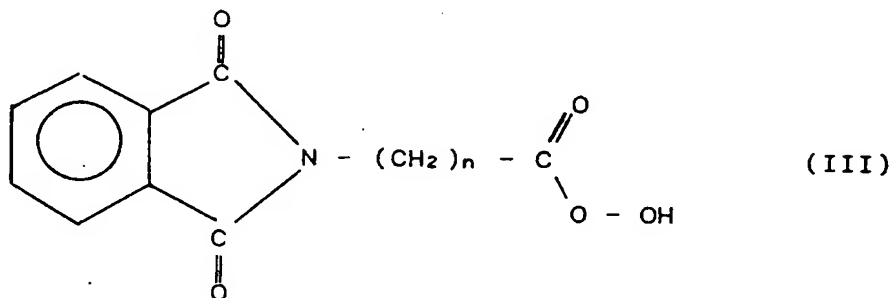
45 (ii) higher than that amount of raw material which stoichiometrically corresponds to the solubility threshold of said peralkanoic acid in said solvent, at the same temperature as defined in above (i) item ;

(C) the solution obtained according to (B) is continuously reacted, at 10-35 °C, with aqueous H₂O₂, in the presence of said strong acid, with the molar feed ratio of said strong acid to the acid to be peroxidized being adjusted at a value smaller than 2, preferably smaller than 1.3, a reaction product constituted by an aqueous phase and an organic phase being thereby obtained;

50 (D) said reaction product is separated into two component single homogeneous phases, said peralkanoic acid and halogenated solvent are recovered from the organic phase and said halogenated solvent is recycled to the dissolving step as described in (B).

- 55
2. The process of claim 1, in which R₁ and R₂ are selected from among H and C₁-C₅ alkyl groups bearing carboxy, percarboxy, hydroxy (OH), nitro (NO₂) or C₁-C₅ alkoxy substituents.

3. Process according to claim 1 or 2, in which said arylimidoperalkanoic acid is selected from among phthalimidoperalkanoic acids of formula (III) and 1,8-naphthaleneimidoperalkanoic acids of formula (IV):



4. The process of claim 3, in which said peralkanoic acid is ϵ -phthalimidoperhexanoic acid ($n = 5$).
5. The process of claim 4, in which the peroxidation takes place inside reactors selected from among static mixers, external-recycle reactors and the reactors of CSTR type, with the number of reactors being of either 1 or 2, and the feed of the reactants being accomplished at one or more successive feed points.
6. The process of claim 4, characterized by the following parameters:
- feed of a solution containing at least 20%, preferably 25%, by weight of PAC in methylene chloride or chloroform;
 - continuous feed of aqueous hydrogen peroxide at 50-90% by weight, preferably 60-80% and with a molar ratio of H_2O_2 (100%) to PAC of from 1.01 to 2, preferably of from 1.01 to 1.6.
7. The process of claim 4, in which the strong acid is selected from among H_2SO_4 , oleum, methanesulfonic acid, trifluoromethanesulfonic acid, fluorosulfonic acid and their mixtures, with the molar ratio of said strong acid to the alkanoic acid to be peroxidized being comprised within the range of from 0.50 to 1.29.
8. The process of claim 7, characterized in that the solution of the acid to be peroxidized in the halogenated solvent is caused to react with hydrogen peroxide, before being brought into contact or during the contact with said strong acid, in the presence of a so-said "solid acid", preferably selected from the group consisting of the cation exchange resins, of strong type, and the zeolites of PENTASIL type in their acidic form, in particular HZSM5 and HZSM11.
9. The process of claim 4, in which the double-phase, reaction product is separated into its component two phases, the organic phase and the aqueous phase, and the residual acidity of the organic phase is removed by a neutralization treatment.
10. The process of claim 9, in which deionized water is added to said aqueous phase and/or to the reaction product before the separation, with the amount of deionized water being of from 0.5 to 1.5 litre per each litre of aqueous phase.

11. The process of claim 9, in which said treatment of neutralization of the organic phase is selected from among:

(a) washing with alkaline solutions, preferably containing NaHCO_3 , NaOH , $\text{Mg}(\text{HCO}_3)_2$ or mixtures thereof;

(b) the flowing through a solid neutralizing bed, preferably selected from the group consisting of NaHCO_3 , Na_2CO_3 , MgCO_3 and mixtures thereof.

12. The process of claim 9, in which any ϵ -phthalimidoperoxhexanoic acid not dissolved in the organic phase is recovered from the aqueous phase by back-extraction with the halogenated reaction solvent.

13. The process of claim 9, in which the organic phase is added with an alkali-earth salt, in particular magnesium sulfate.

14. The process of claim 9, in which the solvent is recovered from the organic phase by cooling, followed by filtration and/or centrifugation.

15. The process of claim 9, in which the solvent is recovered from the organic phase by distillation, with said distillation being selected from among vacuum distillation and steam distillation.

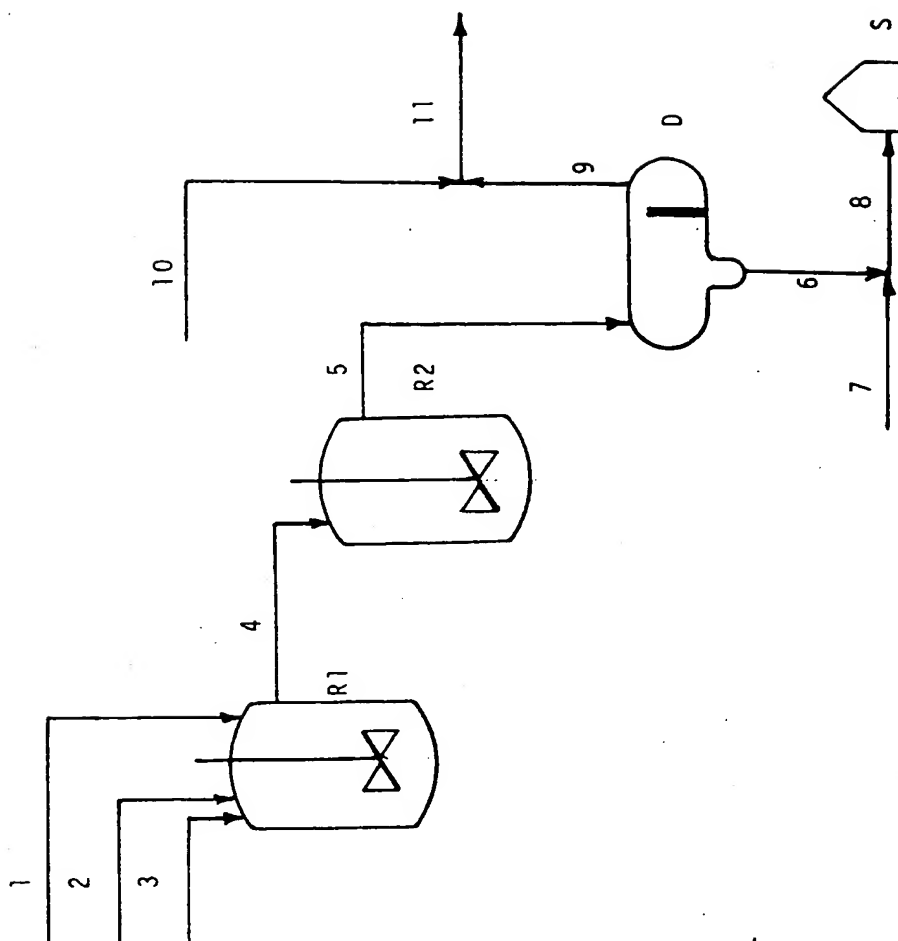


FIG. 1

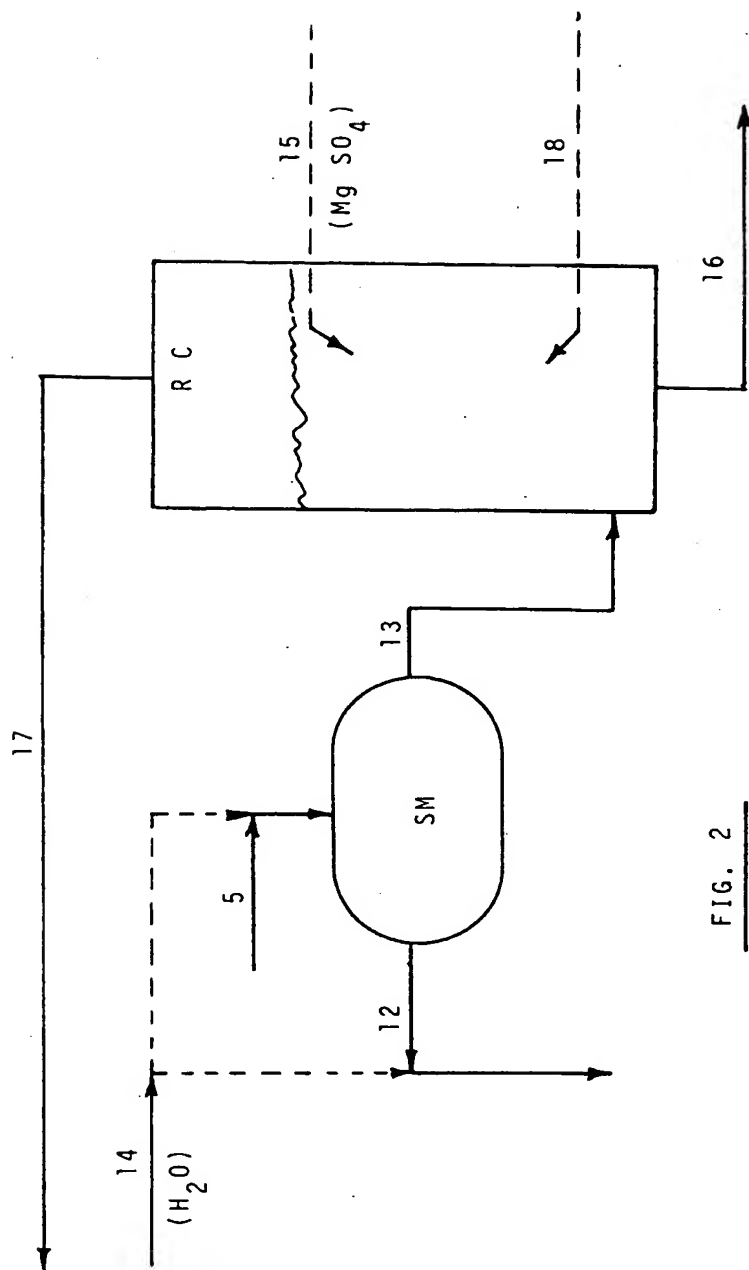


FIG. 2

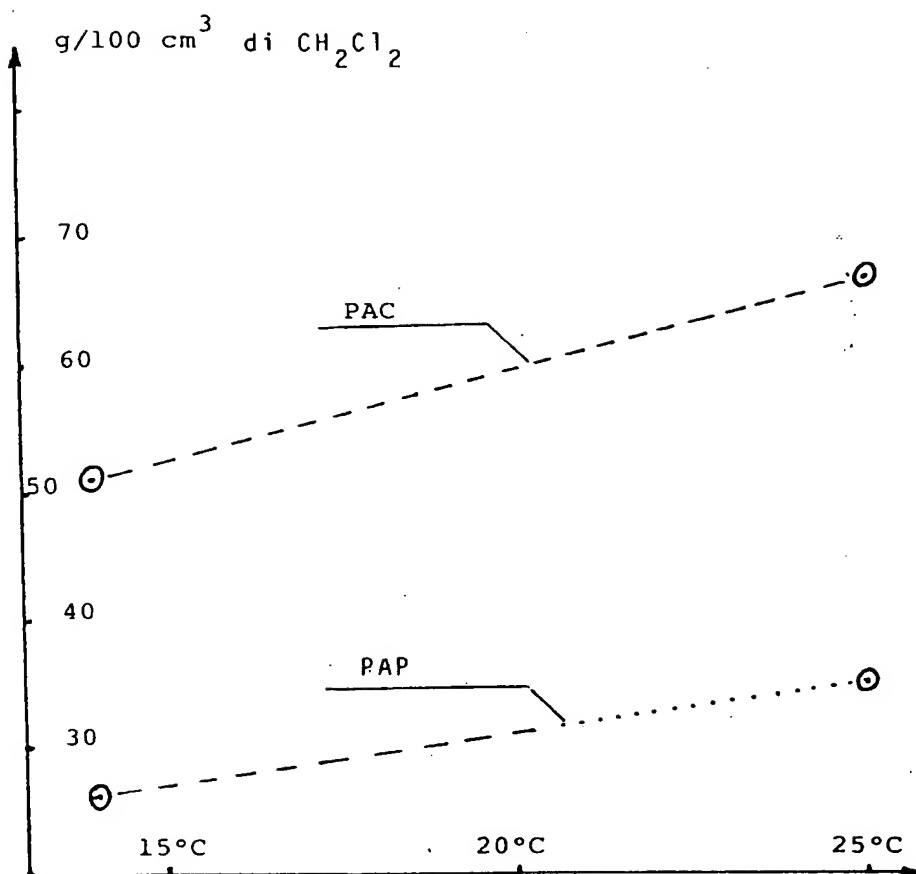


FIG 3



European Patent
Office

EUROPEAN SEARCH REPORT

Application Number

EP 91 12 1453

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
D,A	EP-A-0 325 288 (AUSIMONT S.R.L.) * page 3, line 26 - line 43 * * page 4 - page 5; examples 2,3 * ---	1-3	C07D209/48
D,A	FR-A-2 385 697 (FMC CORPORATION) * page 7 - page 8; example 1 * ---	1	
A	DE-A-3 906 768 (HENKEL KGAA) * page 3 - page 4; examples 1,3 * ---	1	
A	EP-A-0 349 940 (HOECHST AKTIENGESELLSCHAFT) * page 5, line 2 - line 19 * * page 5, line 47 - line 51 * * page 6; example 2 * ---	1-4	
A	WO-A-9 014 336 (INTEROX CHEMICALS LIMITED) * page 25 - page 27; claims 1,2,4,5 * * page 20 - page 21; examples 1,2 * -----	1-4	
			TECHNICAL FIELDS SEARCHED (Int. Cl.5)
			C07D
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 07 FEBRUARY 1992	Examiner FINK D.G.
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons Δ : member of the same patent family, corresponding document			

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